

RESPONSE UNDER 37 C.F.R. 1.196(b)(1)
U.S. Application 09/305,019

REMARKS

Reconsideration and withdrawal of the rejections of record are respectfully requested.

Summary of Status of Amendments and Office Action

Claim 4 to 10 are presently pending in the application, with claim 4 being independent. No amendments to the claims are being made.

In its Decision, the Board of Patent Appeals and Interferences entered new grounds of rejection pursuant to its authority under 37 CFR § 1.196(b). In particular, the Board entered a rejection of claims 4 to 9 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and a rejection of claim 10 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and further in view of Innes et al., U.S. Patent No. 4,891,458.

Response to the Rejection of Claims 4 to 9

Claims 4 to 9 are rejected under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608.

In making this rejection, the Board stated with respect to independent claim 4:

Cheng discloses alkylation and transalkylation processes in which a feed containing an aromatic compound (e.g., benzene) and olefins (e.g., ethylene) is contacted with MCM-56 or beta zeolite catalyst in an alkylation zone to produce monoalkylbenzene products such as ethylbenzene. By-products (e.g., polyalkylbenzene), which are produced in the alkylation step, can be recycled to the alkylation reactor to undergo transalkylation or they can be reacted with additional aromatic feed in a separate reactor in the presence of TEA mordenite catalyst to produce monoalkylbenzene products. The reference discloses that the transalkylation zone is operated in a stoichiometric excess of aromatic compounds. Therefore, the ratio of benzene to polyalkylbenzene is greater than one. (See col. 3, lines 25-27; col. 6, line 2; col. 12, line 10 through col. 14, line 40).

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Examiner's Answer, pages 30-4. See Cheng, column 14, lines 20-44 (noting a preference for performing the transalkylation step in a separate reactor).

It would have been to one of ordinary skill in the art at the time of the invention to have modified the Cheng process to utilize a TEA mordenite catalyst having an average crystal size of < 0.5 micron given EP '608's teaching that improved transalkylation control in the production of ethyl benzene is attributable "to the use of an aluminosilicate catalyst having a crystal size less than about $0.50\ \mu\text{m}$ (0.50 microns)." EP '608, page 3, lines 29-30.

The Board also rejected dependent claims 5 to 9 over the same combination of references. For purposes of response here, Applicants will direct attention to independent claim 4.

Applicants respectfully submit that there is no *prima facie* case of obviousness, i.e., there is no suggestion to combine the teachings of Cheng et al. with those of Kuchenmeister et al. Further, if a *prima facie* case of obviousness has been established, which it has not, then Applicants have rebutted that case with evidence of unexpected results.

Cheng et al, column 13, line 40 to column 14, line 44, teach a two step alkylation process utilizing two stages, wherein an aromatic feed (benzene) and olefin are reacted in the presence of an alkylation catalyst (such as MCM-56) to produce polyalkylated product and then reacting the polyalkylated product and additional aromatic feed (benzene) in the presence of a transalkylation catalyst. The transalkylation catalyst can be a zeolite including a TEA mordenite. Cheng et al., however, do not set forth the particle size of their transalkylation catalyst, nor do they indicate any preference for the use of a TEA mordenite. Rather, at best Cheng et al. indicate that any transalkylation catalyst can be utilized. ①

Kuchenmeister et al, page 2, lines 19 to 44, and the conditions disclosed on page 3, lines 20 and 44, teach a method for transalkylating polyalkylbenzenes in the vapor phase with a catalyst which has a sodium content of less than 50 ppm. At page 3, lines 19 to 26 and lines 19 to 35, Kuchenmeister et al teach that the catalyst is primarily monoclinic

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aluminosilicate, but can contain up to about 40% orthorhombic crystalline structure and has a particle size of less than about 0.5 micron. (V)

Applicants submit that there is no *prima facie* case of obviousness because there is no motivation to combine the teachings of Cheng et al. with those of Kuchenmeister et al. If a *prima facie* case of obviousness has been established, which it has not, then Applicants have rebutted that case with unexpected results.

Applicants submit that there is no suggestion to modify Cheng et al. with Kuchenmeister et al. Cheng et al. broadly teach that any transalkylation catalyst including TEA mordenite may be used as a transalkylation catalyst in the Cheng et al. process. Kuchenmeister et al. teach that their transalkylation catalyst is primarily monoclinic aluminosilicate, but can contain up to about 40% orthorhombic crystalline structure. Applicants submit that Kuchenmeister et al. teach away from the use of a mordenite catalyst, since mordenite has an orthorhombic crystalline structure. In this regard, the Examiner's attention is directed to Meir and Olson, "Atlas of Zeolite Structure Types," Appendix A, page 144 (Butterworth-Heinemann 1992). If the teachings of Kuchenmeister et al. were incorporated into Cheng et al., Kuchenmeister et al. would teach a transalkylation process in the vapor phase using a catalyst having a sodium content less than 40 ppm and having a monoclinic structure. Applicants submit that the mordenite described in Cheng et al. has not been shown to have these properties. It is Applicants' position that the mordenite of Cheng et al. would not have the aforementioned properties. See Meir and Olson publication, *supra*. Further, if Kuchenmeister et al. were combined with Cheng et al., the combination would result in a transalkylation process being conducted in the vapor phase in contrast to the claimed invention which recites that transalkylation is conducted in the liquid phase. (3)

If a *prima facie* case of obviousness has been established, which it has not, Applicants have rebutted that case with unexpected results. In this regard, attention is directed to the Declaration under 37 CFR 1.132, which shows the results of testing comparing a catalyst made in accordance with the present invention and having a particle size of less than 0.5 micron (Example 1 of the Declaration) with two mordenite catalysts having a particle size greater than 5 microns. (4)

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The results of the batch testing show that the inventive Example 1 catalyst has greater activity than a catalyst having a particle size greater than 5 microns. In this regard, the equilibrium limited amount (84%) of diisopropylbenzene was converted into cumene after only 3 hours of reaction time using a catalyst of the present invention, whereas neither the Example 2 catalyst nor the Example 3 catalyst achieved 84% conversion limit even after 7 hours of reaction. This, Applicants submit, demonstrates the unexpected results of the present catalyst. In this regard, the increased activity of the catalyst of the invention is of particular significance because in manufacturing cumene the process can be performed in a shorter time with an increased yield.

For the foregoing reasons, Applicants request that the rejection be withdrawn.

Response to the Rejection of Claim 10

Claim 10 is rejected under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and further in view of Innes et al., U.S. Patent No. 4,891,358.

It is Applicants' position that this rejection fails for the same reasons as given above with respect to the rejection of claims 4 to 9, i.e., there is no *prima facie* case of obviousness because there is no motivation to combine the teachings of Cheng et al. with those of Kuchenmeister et al. If a *prima facie* case of obviousness has been made for the purposes of this rejection, Applicants rely upon the arguments made above with respect to the rejections of claims 4 to 9 and incorporate those reasons herein.

For the foregoing reasons, Applicants request that this rejection be withdrawn.


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CONCLUSION

Applicants respectfully submit that the foregoing arguments place the application in condition for allowance. Allowance of this application is therefore earnestly solicited.

Respectfully submitted,

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PATENT
Docket No. 10151-1**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Patent Application of:)
W. Roth et al)
Serial No. 09/305,019) Examiner: Tam Nguyen
Filed: May 4, 1999) Group Art Unit: 1764
For: ALKYLAROMATICS PRODUCTION)

DECLARATION UNDER 37 C.F.R. 1.132

Commissioner of Patents and Trademarks
Washington, DC 20231

Sir:

I, the undersigned, Dr. Wieslaw J. ROTH, a citizen of United States, whose address is 123 Boundbrook Ct., Sewell, NJ 08080, do solemnly declare, as follows:

1. I have a Ph. D. in Chemistry (Southern Illinois University, Carbondale, IL; 1981) and 15 years experience in the synthesis and characterization of zeolites and catalysts initially at Mobil Research and Development Corporation and, since the 1999 merger between Mobil and Exxon, at ExxonMobil Research and Engineering Company.

2. I am one of the inventors of the above-identified application.

3. The present invention, as recited in claim 4, is directed to a process for producing a monoalkylated aromatic compound comprising the steps of:

- (a) contacting an alkylatable aromatic compound with an alkylating agent in the presence of an alkylation catalyst in an alkylation reactor to provide a product comprising said monoalkylated aromatic compound and a polyalkylated aromatic compound, and then

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- (b) contacting the polyalkylated aromatic compound from step (a) with said alkylatable aromatic compound in the liquid phase and in the presence of a transalkylation catalyst in a transalkylation reactor separate from said alkylation reactor, said transalkylation catalyst comprising TEA-mordenite having an average crystal size of less than 0.5 micron to produce said monoalkylated aromatic compound. [Emphasis added.]

4. It is my understanding that the Board of Patent Appeals and Interferences entered a new ground of rejection pursuant to its authority under 37 CFR § 1.196(b). The Board entered a rejection of claims 4 to 9 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and a rejection of claim 10 under 35 U.S.C. § 103(a) as being obvious over Cheng et al., U.S. Patent No. 5,557,024 in view of Kuchenmeister et al., EP 733,608 and further in view of Innes et al., U.S. Patent No. 4,891,458. In particular, the Board relied upon Cheng et al., column 14, lines 20 to 44, which note a preference for performing a transalkylation step in a separate reactor which can contain TEA mordenite. The Board acknowledged that Cheng et al. do not teach a particle size for the TEA mordenite, but considered that it would have been obvious to modify the Cheng process by utilizing a TEA mordenite catalyst having an average crystal size of less than 0.5 micron, given that Kuchenmeister et al., page 3, lines 29-30, teach that improved transalkylation control in the production of ethyl benzene is attributable "to the use of an aluminosilicate catalyst having a crystal size less than about 0.50 μm (0.50 microns)."

5. The present application at pages 8 and 9 describes four examples, which were conducted under my direction and control.

6. Example 1 of the present application describes the preparation of a TEA-mordenite catalyst having a crystal size of less than 0.5 micron. In particular, a synthesis mixture was prepared which comprised water, precipitated silica, aluminum sulfate

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solution, sodium hydroxide and tetraethylammonium bromide and which had the following molar composition (based alumina = 1):

silica	=	39.7
Na ₂ O	=	7.3
SO ₄	=	2.9
TEA	=	12.3
water	=	370

The synthesis mixture was crystallized at 149° C (300° F) with stirring at 90 RPM for 40-44 hrs. The resultant TEA-mordenite was isolated by filtration, washed and dried and found to have a crystal size by scanning microscopy of less than 0.5 micron.

7. Comparative Example 2 of the present application describes the preparation of a TEA mordenite, which was prepared in accordance with Example 10 of U.S. Patent No. 4,052,472, from a synthesis mixture having the following molar composition: 30 SiO₂, 1 Al₂O₃, 7.5 Na₂O, 6.2 TEA (bromide) and 309 H₂O. The synthesis mixture was crystallized (with stirring at 200 RPM) at 174° C (345° F) for 72 hours. The resultant TEA-mordenite was isolated by filtration, washed and dried and found to have a predominant crystal size by scanning microscopy of greater than 5 microns.

8. Comparative Example 3 of the present application describes the preparation of a TEA mordenite, which was prepared in accordance with Example 8 of U.S. Patent No. 3,766,093, except that sodium aluminate solution containing 25.5% Al₂O₃ and 19.5% Na₂O was used. The synthesis mixture had the following molar composition: 30 SiO₂, 1 Al₂O₃, 4.4 Na₂O, 6.2 TEA (chloride) and 308 H₂O. The synthesis mixture was crystallized (with stirring at 200 RPM) at 171° C (340° F) 138 hours. The resultant TEA-mordenite was isolated by filtration, washed and dried and found to have a predominant crystal size by scanning microscope of greater than 5 microns.

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9. Example 4 of the present application describes the preparation and testing of transalkylation catalysts, which were produced from the zeolite materials produced in Examples 1 to 3. In particular, each zeolite of Examples 1 to 3 was blended with a 35% alumina binder and water and then each blend was extruded into 1/16 inch cylindrical form. The extrudates were then precalcined in nitrogen at 540°C (1000° F) and subsequently ion exchanged with 1 N NH_4NO_3 and calcined in air at 540°C (1000°F) to convert the zeolites to the hydrogen form.

10. The testing was conducted as follows with each of the three catalysts made in paragraph 9. A mixture comprising benzene (150g), diisopropylbenzene (50 g), and 8 g of the transalkylation catalyst produced from the small crystal TEA-mordenite of Example 1 was reacted in a Parr autoclave at 204° C (400° F), 2170kPa (300 psig) and 300 rpm for 7 hours. A small sample of the product was withdrawn from the autoclave at 1, 3, 5, and 7 hours and analyzed by gas chromatography. The catalytic activity is shown in Figure 1, which displays the % conversion of diisopropylbenzene (DIPB) versus time of reaction.

11. The above test was repeated for the catalysts produced from the zeolites of Examples 2 and 3. Figure 2 shows the results of the testing of a transalkylation catalyst produced from the TEA-mordenite of Example 2. Figure 3 shows the results of the testing of a transalkylation catalyst produced from the TEA-mordenite of Example 3. The results of the three tests are summarized in the Table below.

Reaction Time, hr	Example 1 (Invention)	Example 2 (Comparative)	Example 3 (Comparative)
1 hr	54	0	5
3 hrs	84	24	14
5 hrs	84	60	30
7 hrs	84	80	50

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12. The attached results demonstrate that the catalyst of Example 1 is significantly more active than the catalysts of Examples 2 and 3. The reaction between diisopropylbenzene and benzene to produce cumene does not go to completion; but instead is equilibrium limited at a conversion level of 84 %. In the case of the catalyst of Example 1, the 84% conversion limit was reached after 3 hours of reaction. In contrast, neither the Example 2 catalyst nor the Example 3 catalyst achieved the 84% conversion limit even after 7 hours of reaction.

13. I consider that the results of the Example 1 catalyst are unexpected and demonstrate that the Example 1 TEA-mordenite catalyst, which has a particle size less than 0.5 micron, is more active in the transalkylation of diisopropylbenzene with benzene to produce cumene than either of the other TEA-mordenite catalysts, which have a particle size greater than 5 microns. This activity of the Example 1 catalyst is of particular significance because, in the manufacture of cumene, it means that the transalkylation can be performed with a shorter contact and/or with an increased yield of the cumene product.

The undersigned further declares that all statements made herein of his own knowledge are true and that all statements made on information and belief knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity in the above-captioned application or any patent issuing therefrom.

Wieslaw J. Roth

Dr. Wieslaw J. Roth

10/16/2002

Date